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(1) Quantum jumps
(2) Coherent transients
(3) Laser-assisted collisions
(4) Collision-induced resonant structures in laser spectroscopy
(5) Collision kernels and transport coefficients
(6) Interactions with broadband noise

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ANNUAL REPORT (MAR 2)

Title: "Theoretical Studies Relating to the Interaction of Radiation with Matter: Atomic Collision Processes Occurring in the Presence of Radiation Fields"

Supported by: The U.S. Office of Naval Research

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Research has been carried out in the areas of (1) the theory and interpretation of collision-induced resonances, (2) laser-assisted collisions, (3) the interaction of an atomic vapor with broadband light sources, (4) quantum jumps in a two-atom system, (5) "exchange" collision kernels and (6) coherent transient spectroscopy.

1. Theory and Interpretation of Collision-Induced Resonances (P.R.Berman)

In collaboration with G. Grynberg at the Ecole Normale Supérieure in Paris, we have begun a systematic study of so-called pressure-induced extra resonances (PIER). Research into these resonances was stimulated in large part by the work of Bloembergen and coworkers[1], in which the resonances were interpreted as arising from the "destruction of destructive interference" of various density matrix perturbative contributions to the signal. We do not favor this interpretation and have been able to develop an alternative explanation for the resonances which we believe is more attractive from a physical viewpoint. In doing so, we have also discovered new nonlinear optical interactions which give rise to PIER.

Our first paper in this area[2*] analyzed three-level atoms interacting with four incident laser fields. The three levels are in the ladder configuration and two of the fields drive one of the atomic transitions while the other two drive a coupled transition. The upper state population is monitored as a function of the frequency difference of the applied fields. All the features encountered in studies of PIER using the collective emission of four-wave mixing[1] are also found in this system in which an atomic-state population is monitored. Moreover, we are able to interpret the system using a dressed atom approach. The amplitude of the pressure-induced resonance is directly linked to a dressed-state population which vanishes in the absence of collisions. The position and width of the resonance is interpreted in terms of a level crossing between dressed states. Both semiclassical and fully-quantized dressed atom approaches were used.

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* An asterisk indicates that a reprint or preprint of this article has been forwarded to the Scientific Officer with this report. Reprints of articles have been furnished to DTIC with this report. Preprints or reprints of articles are available on request to anyone receiving this report.

In a second paper,[3*] we used a semiclassical dressed atom approach to interpret the PIER that occur in pump-probe absorption and 4-wave mixing. Moreover, we derived a signal for pressure-induced resonances in fluorescence beats. In this approach, the dressing field is taken as the average of two incident fields with a (time-dependent) amplitude. The approach represents a relatively simple calculational tool in which the PIER can be traced to a collision-induced, <u>modulated</u> population of the semi-classical dressed states.

In a third paper,[4*] the semiclassical approach was extended to calculate PIER in (1) four-level atoms interacting with four incident fields (2) three-level atoms (+ continua) which are photoionized by four incident radiation fields and (3) three level atoms in which the PIER is monitored via fluorescence. All these cases have not been analyzed previously, to the best of our knowledge.

In a fourth paper,[5*,6*] we used a quantized field approach to analyze the PIER that are produced via four-wave mixing when three incident fields having frequencies Ω, $\Omega+\delta$, Ω are incident on an ensemble of two-level atoms. This is by far the most difficult system to analyze since the atoms remain in the two-level subsystem after spontaneous emission. We were able to overcome the spontaneous emission problem by defining dressed states in terms of operators rather than number states. With this method, the PIER are linked to the collision-induced creation of modulated dressed-state population operators. In a number-state representation, we still have some problems of interpretation. These problems have led us to reinvestigate the basic assumptions that go into dressed-state theories involving spontaneous emission cascades. Work in this area is continuing.

In all the above cases, we have shown that the vanishing of the PIER in the absence of collisions is intimately related to the conservation of energy. Thus, we have achieved a rather physical picture of the collisional-radiative interaction.

2. Laser-Assisted Collisions (P. Berman)

In collaboration with F. Schuller (Laboratoire de Physique des Lasers - Villetaneuse, France), we have finished a study of final-state polarization produced in laser-assisted collisions.[7*] The results are calculated in the quasistatic-wing, weak excitation field limit for light-induced collisional excitation transfer (LICET). In these reactions, two atoms collide and absorb a photon, taking each atom to a new state. The final state magnetic polarization of one of the colliding atoms is monitored as a function of detuning and serves as a probe of the collisional interaction. Our results are compared with experiment[8] and corresponding theories for optical collisions[9] (in which one of the colliding atoms remains in the same state). The importance of the collision dynamics in the LICET reaction is stressed.

A review of our work on a three-state model of LICET has also appeared.[10*]

3. Broadband Light Sources (V. Finkelstein, P. Berman)

It has recently been appreciated that broadband light sources can serve as a source of sub-picosecond time resolution.[11,12] Rather than the pulse duration, it is the correlation time of the light source that determines the time resolution in certain limits. A photon echo or stimulated photon echo is often used to exploit the inherent high temporal resolution of broadband noise.

The theoretical analysis of this problem brings into play many profound problems, owing to the fact that the same light source interacts with the active medium at least twice. The incident beam is sent into the medium as is its time-delayed replica. Consequently, there is a "memory" of the initial pulse and the problem is non-Markovian in nature. Especially in intense fields, the theory poses formidable problems. Attempts at solutions[13,14] have been rather complicated.

We have made progress towards a simplified theoretical approach to this problem. Using both analytical and numerical techniques, we

have solved the stimulated and photon echo problems in the limiting case of a small time delay between the first two incident pulses. The solution[15*], is based on an "effective-field method" in which the two time-delayed pulses are replaced by totally overlapping (but modified) pulses. With the use of modified overlapping pulses, the problem can be solved using a decorrelation approximation. This method enable us to give a physical explanation of the solution and to give a meaningful comparison to the results for correlated and non-correlated incident fields. In intense fields, the solutions exhibit a temporal structure on the order of the field correlation time, in agreement with experiment.[12]

Our work in this area is continuing, in collaboration with the experimental group at Laboratoire Aimé Cotton, France. We also maintain contacts with Hartmann's group at Columbia.

4. Quantum Jumps in a Two-Atom System (K. Yamada, P. Berman)

We have analyzed the macroscopic quantum jumps that can occur when two identical atoms separated by a distance d are irradiated by a laser of wavelength λ which is nearly resonant with a transition in each of the atoms.[16*] The eigenstates of the two atoms consist of symmetrical and antisymmetrical components. When $d \ll \lambda$, the symmetrical eigenstates decay at rate 2Γ (Γ = decay rate of the excited state of an isolated atom) while the antisymmetrical eigenstate is metastable. Consequently the antisymmetrical state can be used as a "shelving" level to produce quantum jumps. We have worked out the probability distributions for the dark and bright periods assuming both coherent and incoherent pumping of the atoms. Moreover, we have calculated the photon statistics and frequency-resolved photon statistics of the spontaneously emitted light, using a method involving "frequency-resolved delay functions". It may be possible to observe these effects by implanting impurity atoms in a host crystal.

5. "Exchange" Collision Kernels (G. Rogers, P.R. Berman)

In a previous article[17], we examined the relationship between the collision kernels used to analyze experiments in laser spectroscopy and the collision integrals and transport coefficients of kinetic theory. Equations were derived which served to connect the collision kernels and

the collision integrals. The theory has implications for using transport data to reduce the number of free parameters in fitting laser line shapes. It also serves to test phenomenological collision kernels. We are extending this calculation to "exchange" kernels, which give the probability density per unit time that, if atom A enters a collision with velocity $\vec{v}'$, then the colliding partner A' leaves the collision with velocity $\vec{v}$. Such kernels are needed to calculate coefficients of viscosity or thermal conductivity. Our work on this problem is almost complete, but there remain a few wrinkles to iron out which did not appear when studying the "direct" kernels.

6. Coherent Transient Spectroscopy (R. Sung, E. Block, P. Berman)

Our work on an extended-pulse echo has appeared.[18*] The results are in good agreement with the experiment of Yodh *et al*,[19] but our explanation of the results differs from theirs.

Recently we are analyzing some coherent transient data of A. Szabo using ruby as an active medium.[20] There are still many unresolved problems related to the "failure of the optical Bloch equations" for such systems.[21] Szabo's data may help to shed new light on this problem.

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PUBLICATIONS

PAPERS SUBMITTED TO REFEREED JOURNALS
(not yet published)

1. P.R. Berman and G. Grynberg, "Quantized-field approach to pressure-induced resonances," Phys. Rev. A (in press) (additional support from NSF).

2. K. Yamada and P.R. Berman, "Macroscopic quantum jumps from a two-atom system," submitted to Phys. Rev. A (additional support from NSF).

3. V. Finkelstein and P.R. Berman, "Optical coherent transients induced by time-delayed fluctuating pulses. I: three-pulse transients," submitted to Phys. Rev. A (additional support from an NSF International Grant).

4. P.R. Berman, F. Schuller and G. Neinhuis, "Generation of magnetic coherence in light-induced collisional excitation transfer: Weak field, quasistatic-w ng limit," submitted to Phys. Rev. A.

5. G. Grynberg and P.R. Berman, "Pressure-induced extra resonances in nonlinear spectroscopy," submitted to Phys. Rev. A (additional support from NSF).

PAPERS PUBLISHED IN REFEREED JOURNALS

1. P.R. Berman and G. Grynberg, "Theory and interpretation of pressure-induced resonances," Phys. Rev. A39, 570-585 (1989).

2. G. Grynberg and P.R. Berman, "Pressure-induced effects in two-level atoms: New approach and simple physical interpretation," Phys. Rev. A39, 4016-4025 (1989).

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BOOKS (AND SECTIONS THEREOF) SUBMITTED FOR PUBLICATION

1. P.R. Berman and G. Grynberg, "Dressed-atom approach to collision-induced resonances," in Laser Spectroscopy IX, edited by M. Feld, A. Mooradian and J.E. Thomas, to appear.

2. V. Finkelstein, "Excitation into a Quasicontinuum by a Fluctuating Laser Field," in Coherence and Quantum Optics 6, edited by J.H. Eberly, L. Mandel and E. Wolf, to appear

3. C. Finkelstein and P.R. Berman, "Stimulated Photon Echo Induced by Broad-Bandwidth Pulses, in Coherence and Quantum Optics 6, edited by J.H. Eberly, L. Mandel and E. Wolf, to appear.

4. K. Yamada and P.R. Berman, "macroscopic Quantum Jumps from a Two-Atom System," in Coherence and Quantum Optics 6, edited by J.H. Eberly, L. Mandel and E. Wolf, to appear.

BOOKS (AND SECTION'S THEREOF) PUBLISHED

1. P.R. Berman, "Three-State Model for Laser-Assisted Collisions," in Spectral Line Shapes, edited by J. Szudy (Ossolineum, Warsaw, 1989) V.5, pp. 713-732.

INVITED PRESENTATIONS

AT TOPICAL OR SCIENTIFIC/TECHNICAL SOCIETY CONFERENCES

1. P.R. Berman and G. Grynberg, "Dressed-atom approach to collision-induced resonances," Ninth International Laser Spectroscopy Conference, Bretton Woods, NH, June, 1989.

2. P.R. Berman, "Collision-induced coherence," Three lectures at Workshop on "Photon-Assisted Collisions in Atoms and Molecules," Trieste, Italy, February, 1989.

CONTRIBUTED PRESENTATIONS AT

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1. V. Finkelstein, "Excitation into a Quasicontiuuum by a Fluctuating Laser Field," Sixth International Conference on Coherence and Quantum Optics, Rochester, NY, June 1989.

2. V. Finkelstein and P.R. Berman, "Stimulated Photon Echo Induced by Broad-Bandwidth Pulses," Sixth International Conference on Coherence and Quantum Optics, Rochester, NY, June 1989.

3. K. Yamada and P.R. Berman, "Macroscopic Quantum Jumps from a Two-Atom System," Sixth International Conference on Coherence and Quantum Optics, Rochester, NY, June 1989.

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